CONSTITUENT OF NATURAL ORGANIC MATTER (NOM) AND ITS EFFECT IN WATER

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Abstract

Natural organic matter should be carefully considered in terms of its constituent and effect because NOM is complex substances that occur in spatially and seasonally varying concentrations in natural waters. This review presents charteristics of natural organic matter present in water. These compounds mainly include humic substances, carbohydrates, proteins (amino acids), hexosamines, fats, oils, greases, and trace organic compounds (endocrine disrupting chemicals and pharmaceuticals and personal care products).

Keyword: Natural organic matter; water; Characteristics; trace organic matter

1.1 Introduction

Natural organic matter (NOM) originates from the contact of water with dead and living organic matter in the hydrologic cycle, and is a fundamental component of aquatic ecosystems. NOM has important roles in water treatment processes since it produces harmful by-products with oxidants, increases chemical costs and deteriorates product water quality during distribution. Therefore, it is important to characterize NOM from many water supply systems in details.

NOM present in natural waters is constituted of over 1000 organic compounds. Faced with this complexity, traditional analytical and assessment methods use bulk parameters, such as oxidant demand tests or even simpler surrogates like true colour. Indeed, simple methods are affordable and satisfactory in many applications. However, it is increasingly important to better understand the origins, detailed characteristics, and aquatic behaviour of individual NOM constituents to address water quality management and treatment needs more effectively.

1.2 Overview of NOM in Natural Waters

NOM in natural water can be classified into two main groups by size: i) particulate organic carbon (POC) above 0.45 μm and ii) dissolved organic carbon (DOC) below that limit. Both groups include a wide variety of constituents (Figure 1).

POC includes zooplankton, algae, bacteria, and debris organic matter from soil and plants. Usually it represents less than 10% of total NOM, and can be removed easily by solid-liquid separation processes. DOC can impart many adverse effects on water quality so it remains in the focus of research. Figure 2 shows the most significant DOC components in surface waters. The hydrophobic and hydrophilic fractions together often make up the bulk (up to 80%) of DOC. DOC affects essentially all chemical and biological processes in aquatic environments because it controls the surface properties and colloidal stability of particles. It has a stabilising effect, opposite to that of metal ions. The adverse effects of DOC in drinking water supply are precursor for disinfection-by-products (DBPs) formation, exerts coagulant and oxidant demands, fouls adsorbents and membranes, causes aesthetic and corrosion problems, and provides substrate for biomass growth in water distribution networks.

In some cases, the presence of DOC in water can also be beneficial. For example, DOC substances can react with metals and many compounds to reduce availability and toxicity, or some treatment processes implicitly benefit from the physico-chemical effects of DOC on colloids. Further, humic acids can be used as direct means to extract pollutants [3].

2 Characteristics of NOM Components

The predominant fraction of NOM is comprised of complex humic substances, because of the relative abundance of plant materials in the environment [4]. However, many natural biopolymers (polysaccharides, proteins, hexosamines), or various oils, fats, and other components also significantly contribute to aquatic NOM compositions. The nature of the source materials strongly influences the chemical characteristics of the resulting NOM pool. DOC derived from higher plants is rich in aromatic molecules, while microbial sources impart relatively high levels of nitrogen. Geochemical interactions between NOM and porous soil cause selective sorption thus chemical fractionation of organic matter [5].

2.1 Humic Substances (HS)

There are two major pathways by which humic substances can form in nature [6]. Macromolecules, chiefly plant biopolymers, such as lignin or cellulose partially decompose to HS, while the metabolic pathway involves condensation polymerisation reactions [7]. HS represent a broad class of naturally occurring, biogenic, heterogenous organic material. Being polydispersed polyelectrolytes [8] humic substances possess a high degree of molecular irregularity and heterogeneity; hence their chemical definition is impossible [9]. HS consists of multifunctional aromatic components, linked chemically and physically by a variety of aliphatic constituents, and functional groups to form self-similar fractal molecular structures as shown in Figure 3. The acidic nature (typically 8 to 12 meq/g) of HS are attributable to -COOH- and phenolic OH- groups which are unbalanced by the basic functional groups such as – NH_2. HS shows molecular binding tendency towards silica minerals and iron hydroxides. The anionic macromolecular conformations of HS are analogous to those of flexible polyelectrolytes, adjusting to changing pH and ionic strength.

For operational definitions on basis of solubility, the term HS represents three subclasses: 1) humins, 2) humic acids (HA) and 3) fulvic acids (FA). Humins are chiefly allochtonous in origin and washed into aquatic systems by runoff or percolating waters as fine, insoluble particles. HA are intermediates between humins and fulvic acids, soluble above pH 2. Their molecular weight (MW) ranges from 100 to about 100,000 daltons. They contain less aromatic rings and more carboxylic groups than humins. The dominant FA class has lower MW range (600 to 1,000 daltons), has much less aromatic rings but more carboxylic groups, and soluble under all pH conditions. In non-coloured freshwaters and humic-rich waters FA and HA comprise about 40 to 55% and 60 to 80% of DOC, respectively.

HS is natural to the food chain with very low toxicity [10] thus do not pose any health concern. Moreover, HS reduces the chemical stress of aquatic systems indirectly by changing bioconcentrations and reducing metal toxicities [7]. HS is directly involved in particle formation, complexation and binding, metabolic interaction, and hormone-like action. Chlorine reacts with NOM substances like HS to produce trihalometanes (THM), haloacetic acids (HAA) and a host of other halogenated DBPs, some of which are toxic or carcinogenic. HS also react with ozone to produce carbonyl compounds (aldehydes, carboxylic acids) that contribute to biodegradable organic carbon content, thus promoting biofilm formation and microbial regrowth in water distribution systems [4].

2.2 Carbohydrates

The majority of the carbohydrates present in fresh waters originate from terrestrial systems [2]. Plants may release 30% of their organic matter into water as simple carbohydrates or organic acids rich in carbohydrate. Water leachate of vegetable matter is an important source of arabinose, xylose, and glucose in water. Moreover, the majority of aquatic microorganisms and algae also produce carbohydrates, mainly rhamnose, fructose, mannose, and galactose. The carbohydrate content and composition of water varies with source, biological availability, and degree of biodegradation. Polysaccharides are the largest fraction of carbohydrate, followed by sugars bound to HS. Monosaccharides represent about 25%, and the rest is amino sugars, sugar alcohols, sugar acids, and methylated sugars. Table 1 shows typical dissolved concentations in water sources.

Bound carbohydrates predominantly occur in higher molecular mass fractions (above10 kDa) of isolated HS and there are fewer carbohydrates in FA than in HA fractions [12]. In treated drinking water carbohydrates represent 5 to 25% of organic substances. Although some polysaccharides such as lignin are difficult to degrade, in general carbohydrates provide a carbon source to micro and macroorganisms and have important role in biological treatment processes. Carbohydrates do not interfere significantly with traditional treatment technologies but recognised as major foulants in membrane separation processes [13;14;15].

2.3 Amino Acids and Proteins

Proteins are complex, high molecular weight organic compounds that consist of amino acids (AA) joined by peptide bonds. Protein is essential to the structure and function of all living cells and viruses. Thus, in natural waters the majority of AA and proteins come from living cells (extracelluar enzyme), dead cells (debris), whether from animals or plants. Metabolism and catabolism of microorganisms in natural waters produces these materials and HS also consist of about 5-10% protein [16]. The most definitive list on the origin of proteins and AA related to soluble microbial products is provided by Kuo [17]. While over 500 AA have been found in nature, research has been directed towards the 20 essential amino acids with available detailed measuring techniques (Table 2).

Sea and ground waters have the smallest concentrations of AA (around 50 μg/L), followed by oligotrophic lakes and rivers (100 to 300 μg/L), and eutrophic lakes and marshes (600 μg/L). These figures correspond to 2% to 3% of DOC in common drinking water supply sources. Generally, 0.5% is present as dissolved AA, 1% is bound to HS and the rest is bound to other substances [2]. In humic substances HA and FA contain relatively higher amounts of basic and acid AA, respectively.

AA and proteins are potential carbon and nitrogen source for heterotrophs and have received considerable attention because of their importance to protein synthesis, bacterial metabolism, and algal/bacterial interactions. Organic nitrogen can result in the formation of nitrogenous DBPs, which are of health and regulatory concern, and is known to impact membrane processes by fouling, and chlorination by chloramine formation. Nonetheless, organic nitrogen removal across treatment processes is still largely unknown due to the lack of sensitive analytical methods.

2.4 Hexosamines

Hexosamines are derived from terrestial cutin material (waxy substances covering leaves), in water from zooplankton and occur only scarcely in phytoplankton [18;19]. Figure 4 shows the chemical structure of glucosamines. Glucosamine ($C_6H_{14}NO_5$) and galactosamine ($C_6H_{13}NO_5$) are the intermediate between AA and carbohydrates [12]. Up to 8% of HS nitrogen exists as hexosamines. There are HS building blocks that possess a constant ratio of glucosamine to AA, as is known for bacterial cell walls. In surface waters the dominant hexosamine is glucosamine-NH_2, accounting for 53 - 87% of the total. Glucosamine-NH_2 content decreases with depth in surface waters, while galactosamine-NH_2 shows the opposite characteristic. Owing to their general incorporation into structural biopolymer matrices, such as bacterial cell walls and chitinous material (the main ingredient of the shell of insects and crustaceans), hexosamines are relatively resistant to decomposition, whereas AA is labile compared with bulk organic matter [20]. The ratio of amino acids to hexosamines is a good indicator of the degree of organic matter degradation.

Hexosamines are components of chitin and microbial cell wall [21] and thus inherently have low toxicity. N-acetylaminosugars represent a significant percentage of aquatic NOM. Other bacterial polymers, such as polyhydroxybutyrates are almost systematically observed in natural waters; however their relative contribution to aquatic NOM is unknown. Hexosamines are relatively easily removed by clarification treatment and thought to play little role in DBP formation. However, their retainment by ultrafiltration indicates strong membrane fouling properties.

2.5 Fats, Oils, and Greases

Fats, oils and greases (FOG) can be classified into three main classes by origin: 1) vegetable FOG, 2) animal FOG, and 3) mineral oils and derivatives from crude petroleum. Natural FOG consist of a wide variety of organic compounds such as fatty acids, n-alkyl hydrocarbons, n-alkyl acohols, sterols, terpenes, polycyclic hydrocarbons, chlorophyll, fats, waxes, and resins [7]. Among these compounds, differentiation is possible in terms of structure as i) hydrocarbons (aliphatic and aromatic compounds, as detailed by Saliot [22]) or ii) triglycerides (fatty acid and glycerol). The majority of FOG in natural waters exist as fatty acids and glycerines, allowing further classification of FOG in terms of carboxylic acids.

Non-volatile fatty acids (such as steric and palmitic acids) are the degradation products of fats and triglycerides, and are also important constituents of plants. Volatile fatty acids like acetic or valeric acid are short-chain molecules and result from anaerobic metabolism. Hydroxy acids are common intermediates in biochemical pathways, and excreted by algae during photorespiration. Dicarboxylic acids are common degradation products within soils and include oxalic acid and succinic acid. Lignin degradation products are aromatic acids, and occur in water extracts of leaf or conifer needle litter [23]. Their contribution to DOC is presented in Figure 5.

The common man-made hydrocarbons entering natural waters include chlorinated aliphatic and aromatic hydrocarbon (common solvent), polynuclear aromatic hydrocarbons (from combustion sources), saturated and unsaturated alkanes (from waste oil), and water.

FOG removal pathways using microorganisms has been investigated concerning natural waters [24;25], though a much larger body of knowledge exist for specific components, especially in relation to tertiary biological wastewater treatment processes [26;27;28;29;30].

2.6 Trace Organic Compounds

In natural water, there are many different organic compounds that are usually present in very low or trace concentrations. Due to their large numbers, here only some important groups as examples will be mentioned, noting that details are available from Thurman [2] and other works.

Organic sulphur compounds mostly result from decaying algae and can impart taste and odour to water. The concentration of volatile sulphur compounds in raw water ranges from 1 to 350 μg/L and the dominant compound (up to 75%) is dimethylsulfide. Sterols present in cell membranes, have one hydroxyl functional group, thus are only slightly soluble in water. Characteristic levels are from 0.1 to 6.0 μg/L in natural waters. Aldehydes are of biogenic origin resulting from photochemical oxidation, and present in the ng/L to μg/L concentration range. Aliphatic compounds have volatile characteristics hence lower concentrations (10 to 50 ng/L). Aromatic aldehydes originate from lignin and other plant products, are nonvolatile and water soluble. The most common compounds of organic bases are in mononucleotides and nucleic acids. These are fundamental compounds of plant and animal cells, and contribute to DOC concentrations from ng/L to μg/L levels. Alcohols, ketons and ethers contribute a few μg/L portion to DOC in natural waters. Alcohols and ketones can react with chlorine to form harmful compounds. In lake waters usually octanol and butanol are present, while in river water methanol and hexanol are typical. The majority of pigments originate from plants (chlorophyll, xanthophylls, and cartenoids). These compounds degrade relatively fast, hence they usually represent less than 1% of DOC in natural waters.

Recently, Endocrine disrupting chemicals (EDCs) and pharmaceutical and personal chemical products (PPCPs) have been regarded as important emering chemicals. EDCs refer to a class of toxicity in which an endogenous or exogenous chemical has the ability to mimic or block the natural action of endocrine system in animals [31;32]. The ability of certain chemicals to mimic estrogen has been known for decades [33;34;35;36]. However, the discovery that human hormones and a synthetic hormone used as a pharmaceutical for birth control may have been primarily responsible for reproductive impacts in fish below wastewater outfalls stimulated a plethora of new research on EDCs [37;38;39;40]. In response to amendments to the US Safe Drinking Water Act and Food Quality Protection Act mandating comprehensive screening of commercial chemicals for endocrine activity, the US EPA set forth to develop a screening program to evaluate approximately 87,000 chemicals in commerce for which minimal, if any, data are available regarding endocrine-related toxicity. The endocrine system of animals is vast and affects nearly all aspects of metabolism, growth, development, and reproduction. However, the primary classes of EDCs are compounds which mimic or block the natural action of estrogen, androgen, and/or thyroid. PPCPs represent another group of emerging contaminants that can also be detected at ng/L concentrations in water. Collectively, PPCPs are known as PPCPs. Most of the EDCs and PPCPs are more polar than traditional contaminants and the majority have acidic or basic functional groups. These properties, coupled with occurrence at trace levels (i.e. < 1 μg/L) create unique challenges for both analytical detection and removal processes [41]. Molecular structures of several EDC and PPCP compounds can be found in Vanderford et al. [42]. Details of removal of EDC and PPCP compounds in water can be found elsewhere [43;44;45;46]. Table 4 presents general classes of emerging contaminants. The majority of these contaminants are not currently regulated; however, they may be candidates for future regulation once environmental and human health relevance has been established. An interesting characteristic of these contaminants is that they need not persist in the environment to cause negative effects, since their high transformation/removal rates can be compensated for by their continuous introduction into environment [47]. For most of these emerging contaminants, occurrence, risk assessment, and ecotoxicological data are not yet available and it is therefore difficult to predict their actual risk.

Some compounds are known to cause serious problems even when present in very low concentrations in water sources. For instance, some algal products are known toxins, while others, such as geosmin. Pesticides are typically synthetic products thus cannot be properly classified as NOM. However it has been observed that NOM can associate with pesticides and many other organic compounds to form complex and toxic substances [48]. Pesticide concentrations vary according to regions, countries and locations, but may be present in the order of μg/L in many waters.

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Table 1 Concentration of total dissolved carbohydrates in various natural waters [11]

Table 2 Characteristic of essential amino acids

Table 3 Classes of emerging compounds (adapted from Barceló, 2003)

Table 1 Concentration of total dissolved carbohydrates in various natural waters [11]

Natural water		Concentration (μg/L)	% DOC
Ground water		100	1-4
Sea water		250	5-10
River water		500	5-10
Lake	Eutrophic	900	10-15
	Mesotrophic	680	8-12
	Oligotrophic	250	5-10

Table 2 Characteristic of essential amino acids

Name	Nature	IEP	Name	Nature	IEP
Alanine	HP	6.11	Glycine	HL	6.06
Cysteine	HL	5.05	Histidine	Basic	7.60
Aspartic acid	Acidic	2.85	Isoleucine	HP	6.05

Glutamic acid	Acidic	3.15	Lysine	Basic	9.60
Phenylalanine	HP	5.49	Leucine	HP	6.01
Asparagine	HL	5.41	Methionine	HP	5.74
Glutamine	HL	5.65	Proline	HP	6.30
Serine	HL	5.68	Arginine	Basic	10.76
Tryptophan	HP	5.89	Threonine	HL	5.60
Tyrosine	HL	5.64	Valine	HP	6.00

IEP = isoelectric point, HP = hydrophobic, HL= hydrophilic

Table 3 Classes of emerging compounds (adapted from Barceló, 2003)

Compound class	Examples
	Pharmaceuticals
Veterinary and human antibiotics	Trimethoprim, erythromycin
Analgesics and anti-inflammatory drugs	Codein, ibuprofen, acetaminophen, acetylsalicylic acid, diclofenac, fenoprofen
Psychiatric drug	Diazepam
Lipid regulators	Bezafibrate, clofibric acid, fenofibric acid
B -blockers	Metoprolol, propranolol, timolol
X-ray contrast media	Lopromide, iopamidol, diatrizoate
Steroids and hormones (contraceptives)	Estradiol, estrone, estriol, diethylstilbestrol
	Personal care products
Fragrances	Nitro-, polycyclic-, and macrocyclic- musks
Sun-screen agents	Benzophenone, methylbenzylidene camphor
Insect repellents	N,N-dimethyltoluamide (DEET)
Antiseptics	Triclosan, chlorophene
Surfactants and surfactant metabolites	Alkylphenol ethoxylates, alkylphenols (nonylphenol and octylphenol), alkylphenol carboxylates
Flame retardants	Polybrominated diphenyl ethers (PBDEs), Tetrabromo bisphenol A, Tris(2-chloroethyl)phosphate (TCEP)
Industrial additives and agents	Chelating agents (EDTA), aromatic sulfonates
Gasoline additives	Dialkyl ethers, methyl-4-butyl ether (MTBE)
Disinfection by-products	Iodo-THMs, bromoacids, bromoacetonitriles, bromoaldehydes, cyanoformaldehyde, bromate, NDMA

Figure 1 Typical constituents in surface water and their size ranges [1;2]

Figure 2 Main DOC constituents

Figure 3 Generic structure of HS [7]

Figure 4 Structure of glucosamines

Figure 5 Monomeric carboxylic in surface water [23]

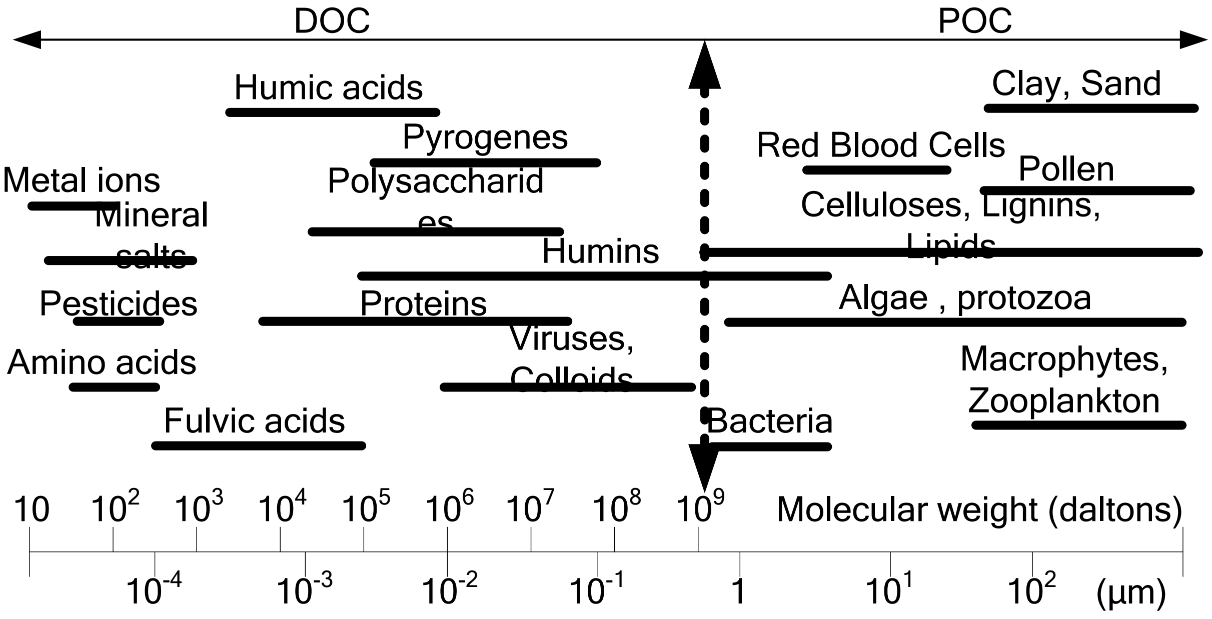


Figure 1 Typical constituents in surface water and their size ranges [1;2]

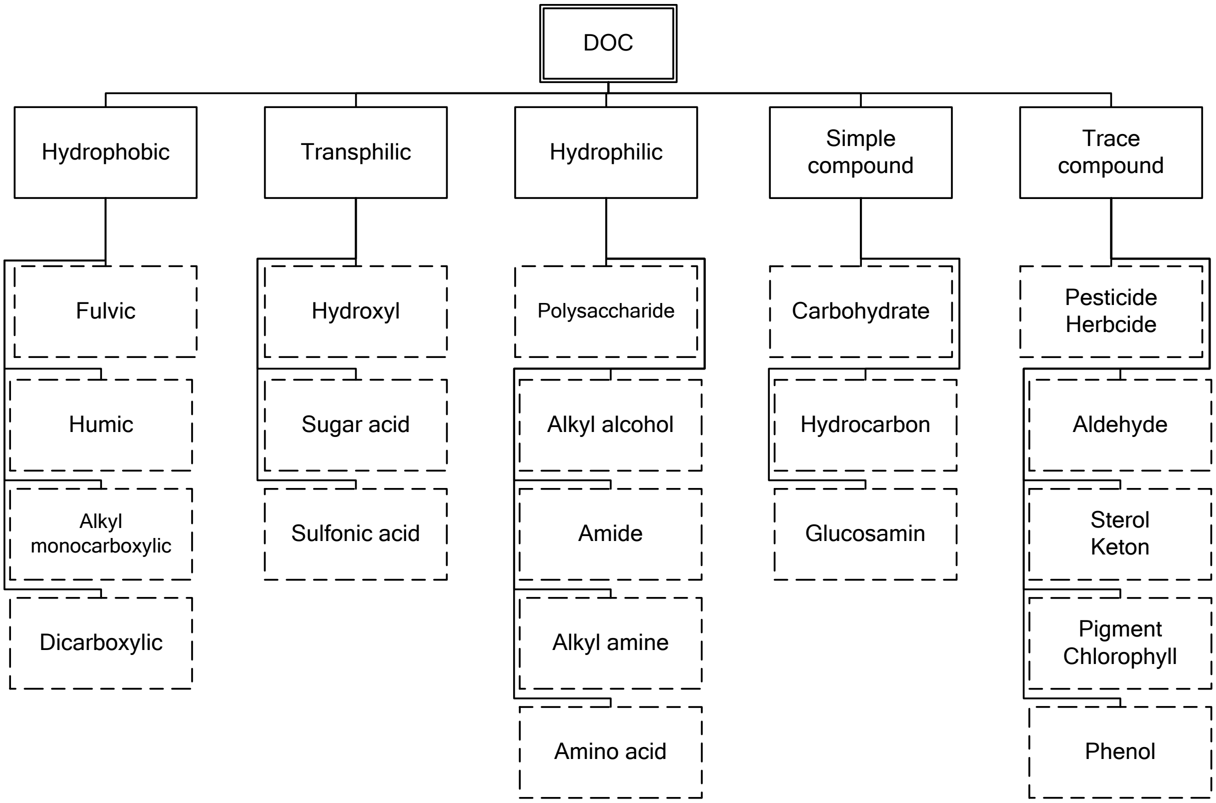


Figure 2 Main DOC constituents

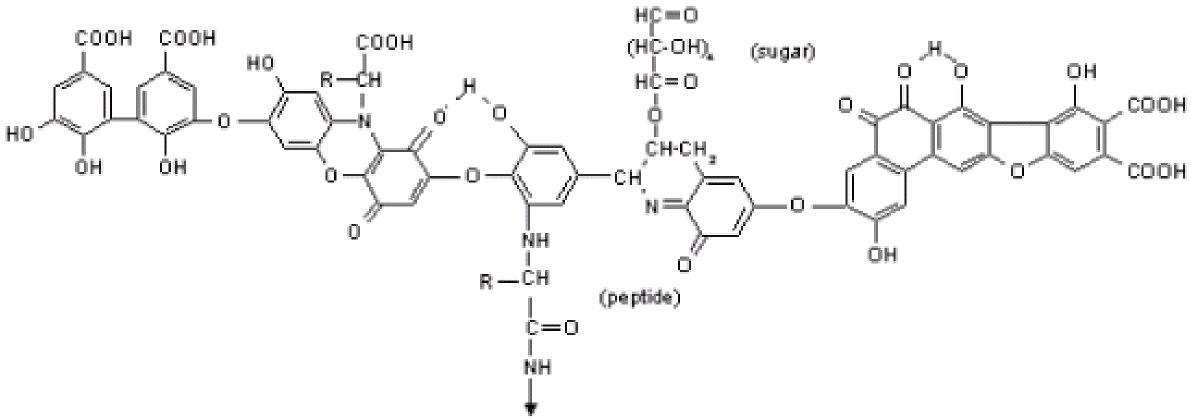
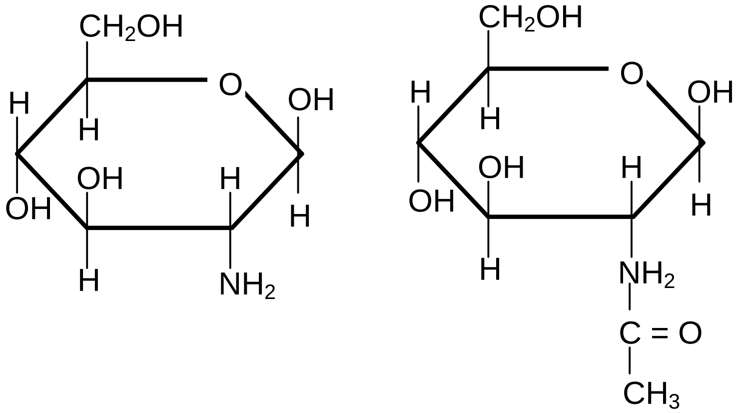


Figure 3 Generic structure of HS [7]



β-D-glucosamine N-acetyl-β-D-glucosamine

Figure 4 Structure of glucosamines

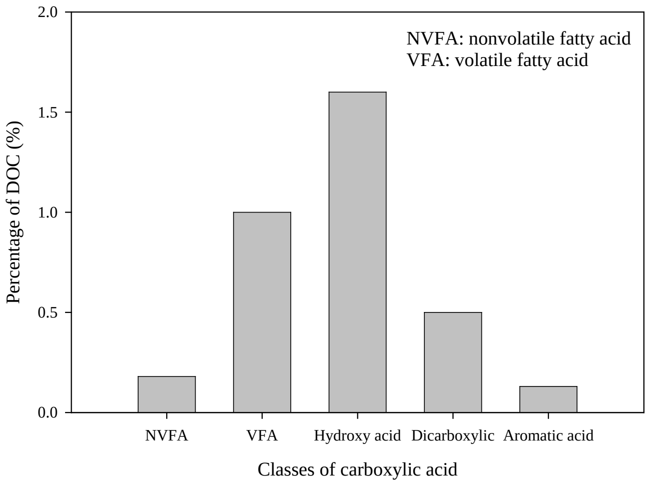


Figure 5 Monomeric carboxylic in surface water [23]